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Ferromagnetic Nano-Particulate and Conductive Mesh Susceptors for Induction-Based Repair of Composites

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Abstract

The Army Research Laboratory is leading research in developing non-autoclave resin curing technologies including a variety of electromagnetic and radiation methods. Recent advances in induction-based electromagnetic bonding have led to several inventions providing enabling technologies for rapid repair of integrated composite structures. Induction-heated bonding of thermoset and thermoplastic polymer matrix composites consists of the heating of a susceptor placed between the parts or of the direct heating of the composite. Induction heating can occur in composite materials due to several mechanisms. The heating obtained by these mechanisms is optimized through the development of various susceptors. The current study examines these susceptor techniques for optimized adaptation to specific Army processing and repair needs. In the case of conductive mesh susceptors, an algorithm was developed which accurately predicts the eddy current production in non-contiguous conductive meshes. The algorithm is then used to optimize the cut patterns in the mesh to provide significant decreases in the susceptor thermal gradients. In the case of ferromagnetic particle-based susceptors, several innovative approaches have been undertaken which serve to optimize heating and to establish a thermally controlled repair process by taking advantage of the Curie temperature. For several repair adhesives studied, a methodology was developed which can be used to accelerate the cure of room temperature curing adhesives for rapid repair. Crosslinking reaction kinetics were developed and employed to determine cure cycles for commercially available epoxy paste adhesives. This paste adhesive was combined with a mesh susceptor for bonding composite adherends. Induction techniques were used to rapidly heat the interface and cure the adhesive consistent with kinetic model predictions.

1. Introduction

The increasing need for lightweight composite structures in the Army necessitates innovative methods of repair and manufacture of complex multi-functional composite systems such as vehicular armor. The Army Research Laboratory is leading research in developing non-autoclave resin curing technologies including a variety of electromagnetic and radiation methods. Recent advances in induction-based electromagnetic heating have led to several Army inventions [1, 2] that provide enabling technologies for rapid repair of integrated composite structures. Induction heating for non-autoclave or non-oven cure and bonding of composites is a novel approach that may reduce manufacturing costs. Supplying the energy needed for curing thermoset resins and adhesives or for thermoplastic bonding is a critical concern in field repair applications and induction heating is an ideal choice. Induction is a non-contact process that can heat geometrically complex parts that are difficult to heat with other bonding methods. Induction-heated bonding of composites consists of the heating of an interlayer susceptor and the subsequent melting, flow, consolidation, and bonding of two thermoplastic-based adherends or the heating, consolidation and cure of a thermosetting adhesive. Induction welding of thermoset composites incorporating a co-cured thermoplastic interlayer is also possible [3]. Susceptors may be resistive, for Joule heating, or magnetic, for hysteresis heating.

Induction-based manufacturing and repair of integrated composite structures depends heavily on the ability to evenly distribute sufficient heat to an adhesive bondline very quickly, in just the right location, and without having direct access to the plane of the repair. While induction bonding of composites has been shown to provide very rapid, non-contact localized heating in the plane of the bondline, it has traditionally been plagued by an inability to sufficiently control the distribution of thermal energy generation throughout the bondline and to control the process temperature. Induction heating occurs when an induction coil is subjected to a high-frequency alternating current and an alternating magnetic field is generated which, in turn, induces thermal generation in a material through either the development of eddy currents in a conductive susceptor or

through hysteresis losses in a ferromagnetic susceptor material. Induction heating can occur in composite materials due to three mechanisms: eddy currents can form in carbon fiber-based composite materials creating dielectric losses in the resin [4-7], eddy currents can form in a conductive mesh susceptor placed at a bondline causing Joule losses [8], and hysteresis can occur in particles embedded in the adhesive causing ferromagnetic domain wall motion and domain realignment losses [9]. The current study is examining all of these susceptor techniques for optimized adaptation to specific Army processing and repair needs.

2. Approach

A technological barrier to the practical application of induction-based processes for adhesive repair is to optimize the heating at the bondline by maximizing the electromagnetic to thermal energy transfer and minimizing in-plane thermal while ensuring that thermal generation in adjacent materials is negligible. In the case of carbon-based composites, a series of models were created and experimentally verified that define the mechanisms and quantification of thermal generation in these systems when subjected to alternating magnetic fields [4-7]. While this type of heating has been demonstrated for volumetric heating of composite sabots, the objective in the repair work is to localize heating at the interface to be repaired. Two parallel courses have been undertaken to address the Joule-loss electrically conductive mesh susceptors and the ferromagnetic particulate-laden adhesive susceptors as described in the following subsections.

2.1 Conductive Mesh Susceptors

Several researchers have conducted tests on the use of metal susceptors (in the form of screens or inserts) and resistive heating for bonding of composites [8-10]. A common problem with metal mesh susceptors subjected to a magnetic field is the resulting non-uniform temperature distributions. Induction coils typically generate non-uniform magnetic fields, though uniform fields can be generated for a few specific coil designs with limited work areas (center of a circular coil). Non-uniform fields can result in temperature gradients exceeding the processing window required for composite heating or bonding. The focus of this work is on developing a new technique, using a metal mesh with specifically designed cut patterns, to generate uniform temperature distributions for non-uniform magnetic fields generated by the coil. The presence of cut patterns in a mesh alters the flow of induced eddy currents and the cut pattern can be optimized to generate uniform temperature distributions.

Mesh density is also an important parameter. If the mesh is too coarse, in-plane temperature gradients between two mesh segments can be large [10]. On the other hand, if the mesh is too fine, there can be poor resin flow across the mesh, resulting in poor bond strengths. Another common problem is the lack of resin material available for flow and bonding and during healing and consolidation. Embedding the mesh in the appropriate polymer system mitigates this problem. In our study, a heat generation model, based on a resistive network type approach, was developed to predict heat generation in a mesh with cut patterns. The alternating magnetic field generated by the induction coil induces eddy currents in the mesh susceptor and the electrical resistance of the mesh material produces Joule-loss heat. The induced electromotive force (emf), in a closed loop in the mesh, can be calculated from:

$$\mathrm{emf} = 2\pi f \mu_o \int_S \mathbf{H} \bullet \mathbf{n}\, dA = 2\pi f \mu_o \sum_{i=1}^{m} \sum_{j=1}^{n} H_{z_{ij}}\, dA$$

where f is the current frequency, μ_0 is permittivity in free space, **n** is a unit vector normal to the mesh surface and H_z the z component of the magnetic field at the surface of the mesh. Since the mesh has a number of closed loops and different loop shapes (in the case of cut patterns), a resistor network type calculation was used to determine the emf's in each segment of the mesh; each carrying an unknown voltage or current. Current conservation laws were applied at each node and, along with the emf equations for each closed loop (induced emf equals the sum of voltages in the loop), a set of linear algebraic equations was obtained and solved for the unknown currents. Knowing the resistance of each segment (from wire geometry and material resistivity) the heat generated in each segment of a mesh box can be calculated. Experimental tests were performed by inductively heating coarse aluminum meshes, with and without cut patterns. A 1 kW Ameritherm induction heater with a 3.75 cm diameter circular coil was used for this purpose (Figure 1). Temperatures in the mesh were measured using infrared thermography.

In a typical induction bonding process, the susceptor is placed between two composite adherends to generate heat at the bondline. In order to have adequate resin flow and consolidation, the susceptor typically contains some resin such as in resin-impregnated metal meshes or an extra layer of resin can be added at the bondline. Consolidation pressure is generally applied by vacuum bagging, though non-metallic rollers may be used for additional pressure.

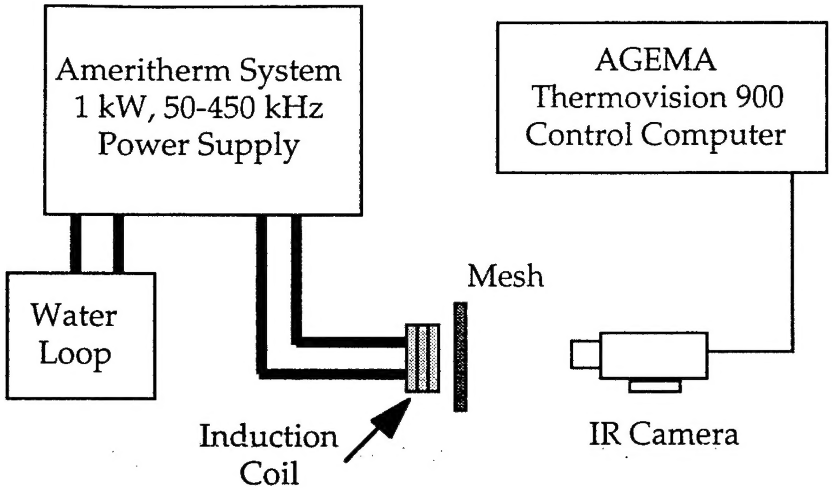


Figure 1. Schematic of induction heating setup.

The coil can be designed to fit complex part shapes and geometries. For large parts, the coil can be moved at a specified velocity to provide necessary heating. The two key requirements of the susceptor are uniform temperature distribution in the susceptor layer and temperature control to avoid thermal degradation of the resin at the bondline and the composite substrate. It is important to note that the use of "uniform temperature distribution" does not necessarily imply the same heat generation throughout the susceptor. Instead, the focus is on maintaining the temperature within the processing window of the composite. Thermal generation gradients may occur within the susceptor and still maintain temperatures within the processing window. Based on bonding models [11, 12] for thermoplastics, this is a key requirement to ensure uniform quality and bond strength.

For the metal mesh susceptor, uniform temperature distribution can be achieved by the presence of cut patterns in the susceptor to redirect current flow paths. The cut patterns will also alter the heat generation and heat transfer mechanisms and an optimized cut pattern can be developed based on a combined heat generation and heat transfer model. Bondline temperature control can be achieved by feedback control based on temperature, input power to the system and coil-susceptor design. To generate uniform temperature distributions within the mesh susceptor, segments of the mesh can be cut to redirect current flow within the mesh. Based on the applied field distribution, preferential heating will occur and cutting segments will force changes in the path of current flow and can optimize heating profiles for optimal adhesive cure development. The current calculation technique described above was adapted for a cut mesh case. Cutting one of the segments forces a "re-direction" of the current loops, resulting in significantly different heat generation in each mesh segment. This model can easily handle larger meshes and more complicated cut patterns and a computer program was developed for this purpose. Some heat generation predictions for different cut-patterns are presented in the results. The model can successfully handle any cutout configuration in the mesh and predict the appropriate induced currents and voltages. Meshes of up to 40 x 40, with many different cut patterns were solved by this model. While the algorithm predicts heat generation, experimental verification requires estimating temperature as the mesh heats up since temperature is the measured quantity. However, because induction heating is a rapid heating process, especially in metals, one can qualitatively compare measured temperature patterns to the predicted heat generation patterns at short times. Integrating the mesh model with a heat transfer model allows temperature comparisons.

2.2 Ferromagnetic Particle-Based Susceptors

In the case of ferromagnetic particle-based susceptors, several innovative approaches have been undertaken which serve to optimize heating and to establish a thermally controlled repair process by taking advantage of the Curie temperature of ferromagnetic materials. The Curie point is the temperature at which the material becomes non-ferromagnetic and no longer generates heat under the influence of an applied alternating electromagnetic field. By using particles with a "dialed-in" Curie temperature that matches the process temperature desired, a self-controlling and self-distributing repair process can be achieved.

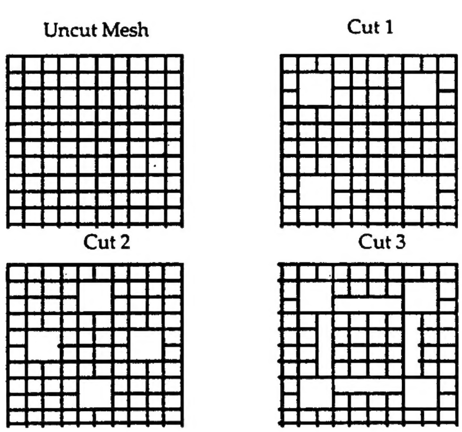
The level of heat generation, the processibility of the particles with the resin, the mechanical performance of the part, and the Curie temperature are all affected by the stoichiometry, particle size, and the frequency of the applied field. Ferrites, while commonly available in a wide variety of Curie temperatures, do not exhibit sufficient domain rotation hysteresis

losses. Hence, our approach has been to investigate the effects of frequency and particle size for a variety of ferromagnetic stoichiometries based on alloys of the transition metals (Fe, Co, Ni) and other elements exhibiting ferromagnetic behavior and have Curie points below room temperature such as the rare earth metals. These materials exhibit very high heating due to domain wall motion and domain rotation. Hysteresis measurements at frequencies ranging from DC to 8 Mhz are being made to locate the resonance peaks of domain wall motion, for various alloys and stoichiometric mixtures, in order to optimize heating rates and minimize the required loading of particles in the adhesives to promote optimal bond strengths. While the stoichiometry has the greatest effect on the Curie temperature, particle size plays an important role in the Curie temperature, the heating rate, and the avoidance of superparamagnetic states. Particle size effects are dictated by ferromagnetic domain structures within the material.

3. Results

Results of the mesh thermal generation model are presented in Figure 2 showing significant improvements in the uniformity of the heat generation between the uncut mesh (indicated by the solid line) and three alternative cut mesh patterns. A 50% reduction in the deviation of thermal generation across the surface of the susceptor is generally possible using the mesh cutting algorithm.

In order to maximize the benefits of accelerated cure of adhesives using induction heating a process window must be established for the adhesives of interest. The process window is used to optimize the bonding process in terms of time and temperature. Issues that dictate the process window include cure kinetics, evolution of exotherms, flow and wetting, and thermally induced residual stresses. Adhesive cure is the most dominant of these issues and must be addressed to determine cure time as a function of temperature as well as ultimate degree of cure. For several repair adhesives studied, a methodology was developed which can be used to accelerate the cure of room temperature curing adhesives for rapid repair. Crosslinking reaction kinetics were developed and employed to determine cure cycles for commercially available epoxy paste adhesives. This paste adhesive was combined with a mesh susceptor for bonding composite adherends. Induction techniques were used to rapidly heat the interface and cure the adhesive. Adhesive taken from the bondline demonstrated full cure at times that were determined from the kinetic models. The use of the model enables prediction of the entire curing process over a wide range of processing temperatures. Initially, however, the prediction of cure time at a specific temperature is of greatest interest to applying induction techniques to accelerate adhesive cure. Here cure time is defined as the amount of time necessary to reach 98% of α_u for each temperature. Figure 3 shows model predictions for cure time compared with the experimentally observed cure times for Araldite AV 8113, a two part epoxy room-temperature-curing paste adhesive from Ciba-Geigy. It was selected because of our prior experience with the system for composite and metal bonding. Also, the manufacturer suggests a 16-hour cure time at room temperature, making it an ideal candidate for accelerated cure studies. While the agreement is not perfect, it does permit an estimate of minimum cure time at each temperature. These values were then used to determine process windows for the induction assisted accelerated cure of this adhesive.



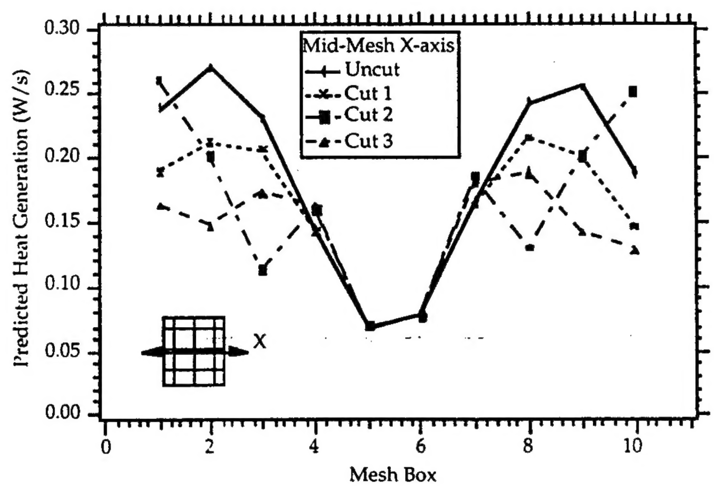


Figure 2. Mesh configurations for cut/uncut case studies and X-axis heat generation for a 10 x 10 square Al mesh: pancake coil.

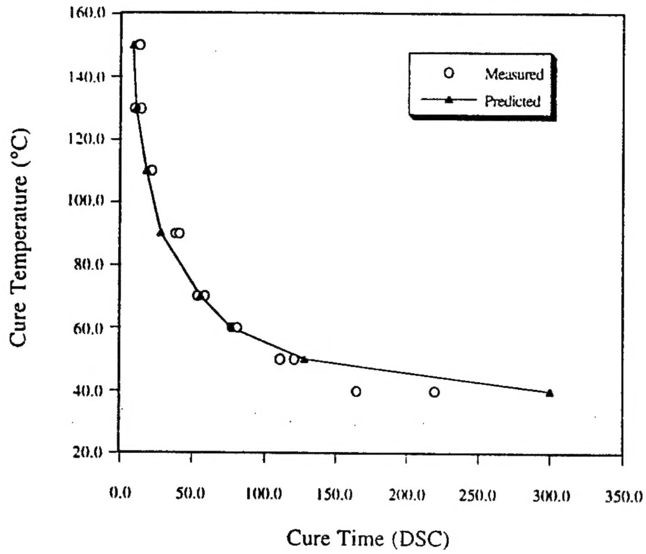


Figure 3. Model predictions for cure time compared to the experimentally observed cure times.

Appropriate cure times for this adhesive can now be selected for any process temperature. This approach was used to select cure cycles for induction heating of the composite adhesive joints. Cure cycles chosen ranged in heating temperature from to 90 °C to 190 °C under vacuum consolidation. Single lap shear specimens were fabricated by induction heating using a stainless steel mesh as the susceptor. An "ear-muff" type induction coil was used and it carried currents between 25-40 A at a frequency of 284 KHz. Typical temperature profiles during induction heating of lap shear specimens are shown in Fig. 4.

A significant advantage to the ferromagnetic particle technique over joule heating techniques is the use of the Curie temperature, T_c, of the ferromagnetic material as a means of automatic temperature control. Choosing materials such that T_c is within the processing window of the polymer allows self-control of the process temperature. An example of this concept is shown in Figure 5. The nickel/polysulphone susceptor shows a very rapid rise to steady-state temperature (~350°C) once the induction coil is activated and then maintains the temperature regardless of any increase in input power. Polysulphone is a commercially used thermoplastic with a manufacturer-recommended processing window between 300 and 360°C, and nickel has a T_c of 354°C, resulting in an ideal combination. In the study of the particle size effects, single-domain particles (~5-10 nm) were observed to exhibit less heating than multi-domain particles. However, over a certain threshold (~10-50 nm), heating drops off considerably due to the statistical probability of the existence of preferentially oriented domains within the preponderance of particles. Heating, defined in part by the area of the hysteresis loop, is largely affected by the rotation of domains after domain wall motion is complete.

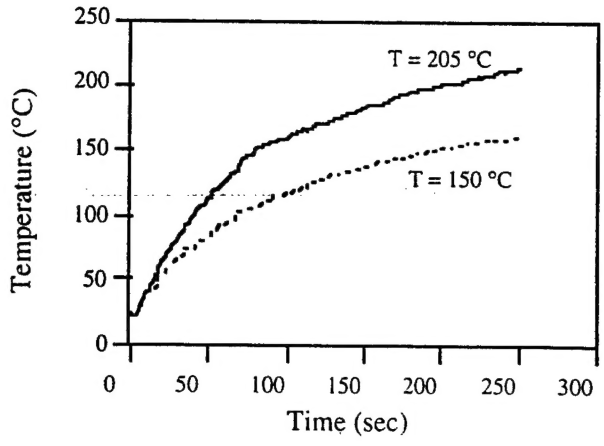


Figure 4. Typical temperature profiles for induction heated adhesive joints.

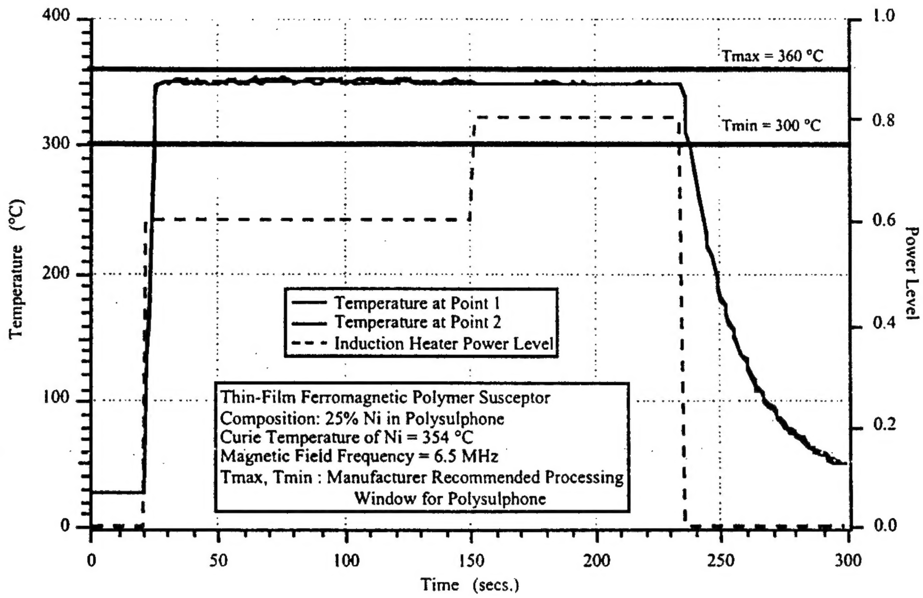


Figure 5. Plot of temperature and applied power level with time demonstrating Curie temperature controllability.

4. Conclusions

New materials and design concepts for conductive mesh and ferromagnetic nanoparticulate susceptors offer the potential to eliminate temperature gradients during induction-based repair of composites. A mesh design algorithm successfully reduced in-plane thermal gradients by a factor of two. High hysteresis-loss ferromagnetic alloys have been fabricated and characterized for Curie temperature process control. Particle size effects indicate low heating for single-domain particles with an optimal maximum for particles with several domains (~10-50 nm). Mesh and particulate susceptors were successfully used to accelerate cure of epoxy paste adhesives. Cure times were reduced from days to minutes for 98% cure.

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